In situ study of reversible formation of self-organized Ge nanostructures on highly miscut Si (001) substrates using GISAXS and GID

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The objective of the experiments was to map out the reversible formation of one dimensional Ge nanoripples on highly miscut Si substrates using in situ grazing incidence x-ray scattering and reciprocal space mapping at the BM32 beamline at ESRF, where SiGe heteroepitaxial growth and GISAXS and GID can be performed in situ at the same time. The goal was to determine the dynamics of the morphological phase transitions during deposition and thermal cycling, and to characterize the evolution of sizes and shapes and the degree of lateral ordering as a function of Ge coverage and substrate temperature. This will allow to shed light on the fundamental nature and mechanisms of this unique self-organized nanopattern formation process.

In our experiments, we used two different Si substrate orientations, namely high-indexed Si (1 1 10) and 4° miscut Si (100) with the miscut in the [100] direction. Whereas the Ge ripples formed on Si(1 1 10) consist of two symmetrical {105} facets with a ripple period to Ge thickness ratio of ≈4nm/ML, the ripples formed on the 4° miscut Si substrates consist of a (105) and a (001) facet with a period ratio of 6.25nm/ML. The Si substrates were chemically cleaned in Linz and transported in DI water. For the oxide removal the samples were flashed for 2 min at 900°C in the BM32 growth chamber and subsequently Ge was deposited with incremental steps at temperatures around 550°C up to different coverages ranging from 1 to 7 monolayers. During deposition GISAXS maps were recorded as exemplified in Fig. 1. The maps were recorded parallel as well as perpendicular to the miscut direction. After each incremental growth step the samples thermally cycled between 350° - 600°C during which GISAX maps were recorded. During these cycles, lateral satellite streaks appeared and disappeared in the GISAX maps signifying the reversible formation and dissolution of Ge nanoripples on the surface as illustrated by Fig. 1 for Ge on Si (1110) and in Fig. 2 for Ge on 4° miscut Si (100). At selected coverages and temperature GID reciprocal space maps were recorded in the vicinity of the (220) and (400) Bragg reflections. As listed in Tab. 1, in total 9 growth runs and 10 annealing cycles were performed in order to determine the critical phase transition temperature and ripple period as a function of Ge coverage.

Table 1: Experimental overview

#	Sample	Experiment
A	Si (001)	Ge rate was measured with RHEED oscillations
B	Si (1 1 10)	Ge deposition (The Ge shutter was not closed properly.)
C	Si (1 1 10)	3, 3.25, 3.5, 3.75, 4, 4.25, 8.95ML Ge; thermal cycles
D	Si (001), 4° miscut in [100]	3, 3.5, 4, 6ML Ge; thermal cycles
E	Si (001), 4° miscut in [100]	10ML Ge growth at T=540°C; GISAXS was recorded during growth
F	Si (001), 4° miscut in [100]	4, 4.3, 4.6, 4.9ML Ge; thermal cycles (350-620°C)
G	Si (001), 4° miscut in [100]	3, 3.3, 3.6, 4ML Ge + 2ML Si + 0.5ML Ge; thermal cycles
H	Si (1 1 10)	3, 3.3, 3.6, 4, 4.3, 4.6, 4.9ML Ge; thermal cycles (300-600°C)
I	Si (1 1 10)	4.3ML Ge; thermal cycles with diff. rates (3, 6, 12°C/min)

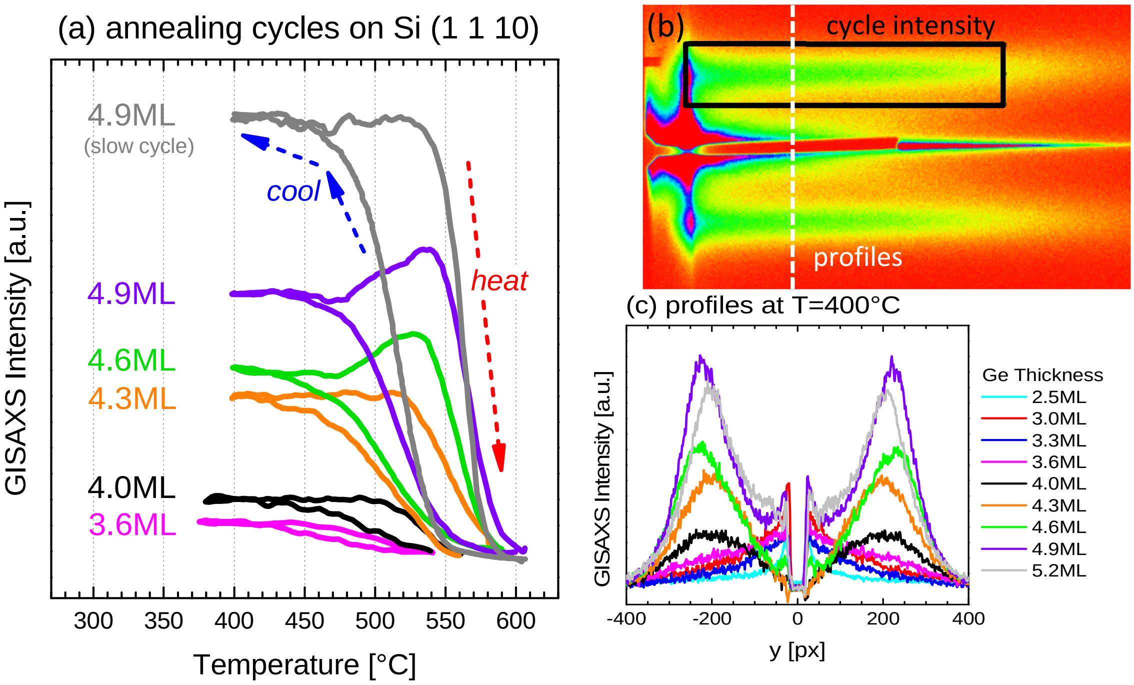

Figure 1: In situ GISAXS experiments for Ge growth and temperature cycling on Si(1 1 10) substrates. In **(a)** the satellite intensity as indicated in (b) is plotted as a function of temperature during thermal cycling between 380°C and 610°C at fixed Ge coverages of Θ_{Ge} between 3.6 and 4.9ML. The cycling rate was *r_T=12°C/min* except for the one slow cycle for 4.9 ML (grey) where *r_T=3°C/min*. High intensity evidences the ripple phase, low intensity the 2D surface without corrugation. The transition between these phases depends on the Ge coverage and shifts from ≈490°C *(3.6ML Ge)* to ≈520°C *(4.9ML Ge)*. **(b)** shows the recorded GISAXS map of 4.9ML Ge after one full cycle (*r_T=3°C/min*) at T=400°C. **(c)** GISAXS profiles, as marked in (b), as a function of Ge coverage after subsequent cooldown (*r_T=12°C/min*) to T=400°C. The average ripple period obtained from the satellite peak positions increases with increasing coverage.

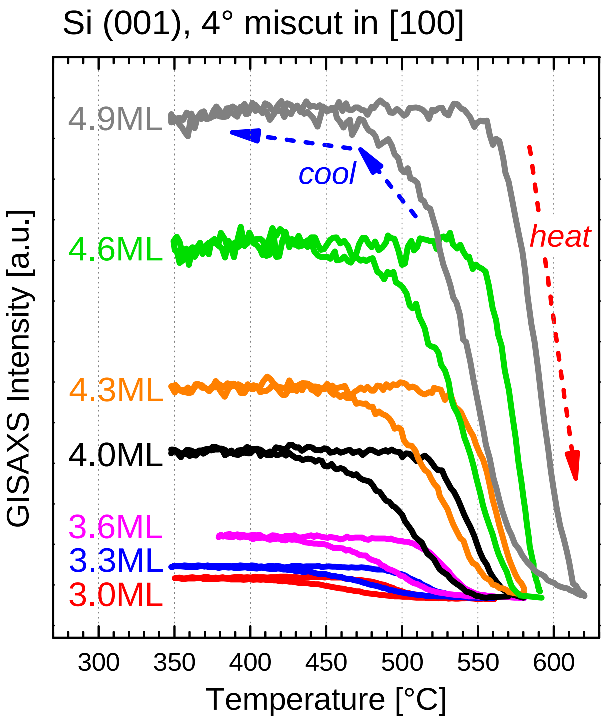

Figure 2: In situ GISAXS experiments for temperature cycling of Ge on Si(001) with 4° miscut in [100] direction. Reversible ripple formation evidenced by the GISAXS satellite intensity curves recorded during cycling between 350°C and 620°C *(r_T=12°C/min)* at fixed Ge coverage of Θ_{Ge} = 3.0 - 4.9ML. The ripple disappearance temperature depends on the characteristic ripple size and shifts from ≈490°C *(3.0ML Ge)* to ≈580°C *(4.9ML Ge)*. (sample F: 4-4.9ML, sample G:3-3.6ML)